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Formation of Very Thin Epitaxial Al_2O_3 Pre-Layer with Very Smooth Surface on Si(111) Using Protective Oxide Layer

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1. Introduction

Epitaxial growth of a single crystalline insulating film on Si has useful applications such as, Si on insulator (SOI), optoelectronic integrated circuits (OEIC's), tunneling and other novel devices [1]. But these devices require a high quality ultrathin and uniform insulating layer. We have proposed mixed source molecular beam epitaxy (Al-N_2O MBE) to grow Al_2O_3 (111) in Si (111) epitaxially [2]. To obtain high quality MBE films, cleaning of the silicon substrate surface is most important. Ishizaka et al [3] reported low temperature thermal cleaning, which involves of the formation of a protective contamination-free thin oxide layer on the surface using a chemical solution and evaporation of this layer in UHV condition at a temperature lower than 800°C. Using this surface cleaning technique, the Si-MBE technique has been recently developed to obtain epitaxial films with low defect density [4]. However, there remain problems such as the surface roughness produced by the holes and SiO_x clusters after thermal decomposition [5,6]. In addition, it is difficult to obtain such a ultrathin and uniform Al_2O_3 layer reproducibly and controllably because of the reaction of Si surface by N_2O gas during the initial Al_2O_3 growth stage.

In view of the above mentioned problems, for the first time, we have examined use of protective thin oxide film as the Al_2O_3 pre-layer, in the succeeding growth of Al_2O_3 films. The Al_2O_3 pre-layer was formed by the Al layer deposited on protective thin oxide layer, followed by annealing. This method is expected to be useful for the prevention of reaction likely to occur during the initial heteroepitaxial growth stage.

2. Experiment

In order to form the Al_2O_3 pre-layer and to grow Al_2O_3 epitaxially, we have used the apparatus consisting of an Al-N_2O MBE growth chamber, a Al deposition chamber, and an analyzer chamber. The Al-N_2O MBE growth chamber is equipped with an Al Knudsen cell and N_2O gas for Al_2O_3 growth, and the Al deposition chamber is fitted with Al Knudsen cell for Al deposition. Epitaxial films were analyzed by in-situ RHEED and XPS techniques. Si substrate used in the present experiments was Si (111) wafer which was pretreated by the Shiraki cleaning method [3]. In order to form Al_2O_3 pre-layer, the thin Al layer was deposited on the thin oxide layer in the Al deposition chamber. Subsequently it was subjected to annealing at 800°C. The epitaxial growth of Al_2O_3 was carried out in the pressure N_2O gas maintained at a constant pressure of 3×10^{-2}Pa. Al Knudsen cell was maintained at 1100°C with an Al flux of ~15 Å/min. The growth temperature was varied from 600°C to 900°C. Crystalline quality and composition of the grown films were studied by in-situ RHEED and XPS techniques, respectively. AFM was also used to study the surface morphology of the grown epitaxial films.

3. Results and discussion

Figures 1(a) and 1(b) show the RHEED patterns obtained after annealing the thin oxide film deposited by using very thin Al layer (i.e. after formation of Al_2O_3 pre-layer). The RHEED photographs show two kinds of patterns: one from a (111) Si substrate and another from a (111) Al_2O_3 epitaxial film. These RHEED patterns indicate the orientation relationship of Al_2O_3(111)//Si(111) with Al_2O_3 <110>//Si<110> and Al_2O_3<112>//Si<112>, corresponding to a lattice mismatch of 2.4% [2].

The AFM image obtained after thermal decomposition of an protective oxide layer on the Si (111) surface is shown in Fig. 2(a). Annealing was carried out at 900°C for 30min. The thickness of the protective oxide film was estimated to be 1.0 nm from the XPS measurement. It is believed

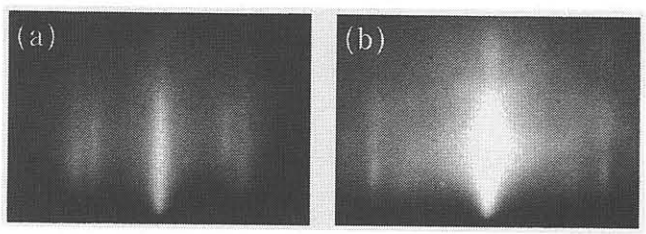

Fig. 1. RHEED patterns observed along (a) <110> and (b) <112> azimuth of the sample formed Al_2O_3 pre-layer

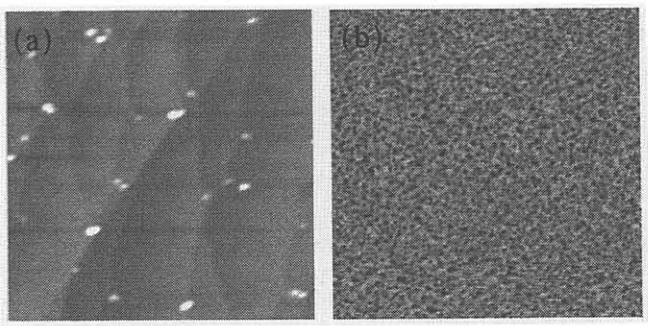

Fig. 2. AFM images (1 μm x 1 μm) after thermal decoposition (a) and after formation of Al_2O_3 pre-layer (b)

that the bright spots is due to SiO_x clusters which are 10 nm in height formed during the thermal annealing process. It is expected that SiO species are desorbed from the surface at high temperature, but it may be possible that these species might have migrated over the surface to form clusters of SiO_x which stay on the surface. In fact, we could not find either a residual oxide layer or contamination by XPS measurement, and a clear 7x7 RHEED pattern was observed over the entire surface. The AFM images obtained after annealing the protective oxide film deposited by very thin Al layer is shown in Fig. 2(b). Smooth surface with an RMS value of 2.7 Å and the Z range for film are comparable and the resulting surface topography reflects the starting Si surface structure without SiO_x clusters.

Before and after the formation of Al_2O_3 pre-layer, the substrate was analyzed using in-situ XPS measurement and the data recorded are shown in Fig. 3. XPS signals corresponding to Si-Si bonds and Si-O bonds are observed from the surface deposited with a protective oxide layer as shown in Fig. 3(a). However, Si-O bonds were not

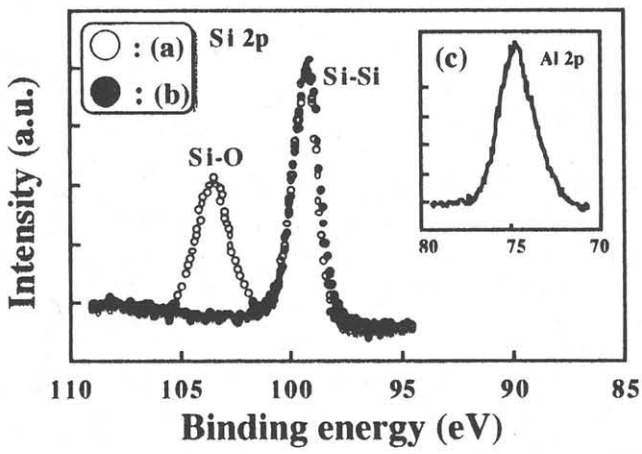

Fig. 3. XPS spectra (a) Si 2p from the sample deposited with protective oxide film, (b) Si 2p, (c) Al 2p, after formation of Al_2O_3 pre-layer.

observed after the formation of Al_2O_3 pre-layer. On the other hand, the intensity of Si signals obtained from the Si substrate after the formation Al_2O_3 pre-layer was similar to that of Si signals obtained from the Si substrate deposited with a protective oxide layer. This indicates that uniform and thin Al_2O_3 pre-layer was formed on the Si substrate. The Al 2p spectra obtained from Al_2O_3 pre-layer as can be seen in Fig. 3 (c) corresponded to the binding energy of Al-O and not to that of Al-Al bonds .

The AFM images of grown Al_2O_3 films with and without Al_2O_3 pre-layer are shown in Fig. 4. The surface morphologies are much different in these images. In case of the growth with pre-layer has a very smooth surface with Z range of ~3 nm. However, the surface grown without Al_2O_3 pre-layer has numerous convex structures due to SiO_x cluster with Z range of ~10 nm.

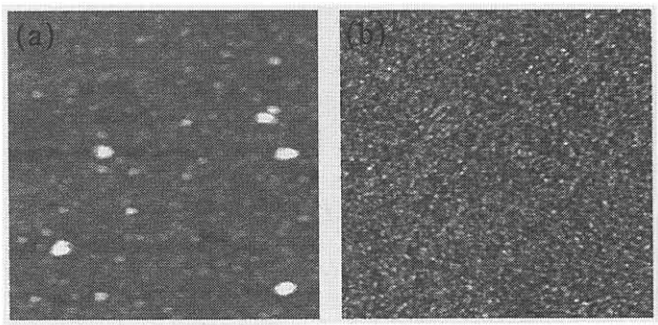

Fig. 4. AFM images (1 μm x 1 μm) of grown Al_2O_3 films (20Å) without pre-layer (a) and with pre-layer (b)

4. Conclusions

In essence, Al_2O_3 pre-layer was successfully grown on Si substrate by annealing process of thin oxide film deposited using a thin Al layer. The very smooth Al_2O_3 (111) film was epitaxially grown on Si (111) substrate using Al_2O_3 pre-layer. It was confirmed that the Al_2O_3 pre-layer was effective in improving the surface morphology of the very thin Al_2O_3 films.

References

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